



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Application of : Jane C. Cheng, et al.
Serial No. : 08/853,007
Filing Date : May 8, 1997
Title : PROCESS FOR PREPARING SHORT CHAIN ALKYL AROMATIC COMPOUNDS
Group Art Unit : 1764
Examiner : B. Yildirim

APPEAL BRIEF UNDER 37 C.F.R. § 1.192

Assistant Commissioner of Patents
Washington, DC 20231

Sir:

Responsive to the Final Rejection mailed December 3, 1998 as to the above-referenced application, a Notice of Appeal having been filed on May 26, 1999, the period for filing the Appeal Brief having been extended by one month by a petition filed concurrently herewith, Appellants submit the following Appeal Brief in triplicate.

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(1) Real Party in Interest.

Mobil Corporation is the real party in interest in this appeal.

(2) Related Appeals and Interferences

There are no other appeals or interferences known to Appellants, Appellants' legal representative, or assignee which will directly affect or be affected by or have a bearing on the Honorable Board's decision in this appeal.

(3) Status of Claims

Original claims 1-10 remain pending in this application, and are all subject to this appeal.

(4) Status of Amendments

Appellants filed no amendments in response to the Final Office Action. A request for reconsideration was filed on April 2, 1999, and was considered by the Examiner; however, the Examiner determined that it did not place the application in condition for allowance. See the Advisory Action, mailed March 28, 1999.

(5) Summary of the Invention

The present claimed invention is directed to a process for preparing short chain alkyl aromatic compounds. At least one aromatic compound is contacted, under alkylation reaction conditions and in the presence of an alkylation catalyst, with at least one alkylating agent possessing an aliphatic group having from 1 to 5 carbon atoms. The

alkylating agent may be an olefin having from 2 to 5 carbon atoms, e.g., propylene or ethylene. The aromatic compound may be, e.g., benzene. The resulting product is an aromatic compound possessing at least one alkyl group derived from the alkylating agent. The alkylation catalyst comprises a binder-free molecular sieve having an X-ray diffraction pattern including the lines set forth in Tables A, B, C, and D at pages 5-7 of the specification.

Additionally, the present invention relates to a process for preparing short chain alkyl aromatic compounds by transalkylation. At least one aromatic compound is contacted, under transalkylation conditions and in the presence of a transalkylation catalyst, with at least one transalkylating agent having two or more aliphatic groups, each having from one to five carbon atoms. The aromatic compound may be, for example, benzene. The transalkylating agent may be, e.g., polyisopropylbenzene or polyethylbenzene. The transalkylation catalyst comprises a binder-free molecular sieve having an X-ray diffraction pattern which includes the lines set forth in Tables A, B, C, and D of the specification.

(6) Issues

There are two issues presented for appeal:

- I. Whether the rejection under 35 U.S.C. § 102(b) is proper when the cited reference does not disclose the claimed alkylation/transalkylation process with a catalyst comprising a binder-free molecular sieve?

II. Whether the rejection under 35 U.S.C. § 103(a) is proper and, if so, whether it has been overcome by the evidence of unexpectedly superior results in the specification?

(7) Grouping of Claims

Claims 1-10 should not stand or fall together, and should be grouped as follows:

1. Claims 1-6, 8, and 10 should stand or fall together.
2. Claims 7 and 9 should stand or fall together, as they are separately patentable.

As will be discussed in the Argument section, and pursuant to 37 C.F.R. § 1.192(c), claims 7 and 9 are separately patentable because the lone reference cited by the Examiner does not teach or suggest, nor does the Examiner allege it teaches or suggests, any type of transalkylation process.

(8) Argument

I. The rejection under 35 U.S.C. § 102(b) should be reversed.

Claims 1-10 are rejected under 35 U.S.C. § 102(b) as allegedly being anticipated by U.S. Patent No. 4,992,606 to Kushnerick et al. The Examiner takes the position that the invention is disclosed in Kushnerick et al. because although the reference allegedly teaches that “it may be desirable” or “it is desirable” to include a binder, the absence of a binder is also within the reference teachings. Final Office Action at page 2. Appellants respectfully submit this rejection should be reversed.

A. Kushnerick et al. Fails to Disclose the Alkylation Process of Claims 1-6, 8, and 10.

Kushnerick et al. fails to teach the claimed process for producing alkyl aromatic compounds by alkylation, wherein the catalyst is a binder-free molecular sieve. As the Federal Circuit ruled in Hybritech, in order to properly anticipate a claim the reference must contain all of the elements of the claim. Hybritech Inc. v. Monoclonal Antibodies, Inc., 231 U.S.P.Q. 81, 90 (Fed. Cir. 1986). The reference does not, however, contain all the elements of the claimed invention.

The Examiner's assertions to the contrary, Kushnerick et al. does not actually disclose an alkylation process using a catalyst comprising a binder-free molecular sieve. The Examiner asserts binder-free molecular sieves are within the teachings of the reference. But Kushnerick et al. actually teaches that

inactive materials may be used which serve as diluents to control the amount of conversion, so that alkylation products can be obtained economically and orderly without employing other means for controlling the rate of reaction.
Col. 9, lines 28-32.

The reference further states that these inactive materials

may be incorporated into naturally occurring clays, e.g., bentonite and kaolin, to improve the crush strength of the catalyst under commercial alkylating operating conditions.
Col. 9, lines 33-35.

Accordingly, it can be seen from the reference passage relied upon by the Examiner that the catalysts envisioned by Kushnerick et al. relate to crystalline materials in combination with inactive materials (such as metal oxides, col. 9, line 21), with said inactive materials being further incorporated into binders. Indeed, an alkylation example in Kushnerick et

al. (Example 15, columns 15 and 16) teaches a catalyst which contains a binder. The present claims, by contrast, specifically recite binder-free catalysts, which means a synthetic porous crystalline material which is free or substantially free of binder materials such as clays or metal oxides. Specification at page 11, lines 32-34.

The reference teaches that it is preferable to include a binder with the catalysts disclosed therein, and the only alkylation examples are directed to a catalyst with a binder. A proper anticipatory reference must put one of ordinary skill in the art in immediate possession of the claimed invention. At most, this reference would put one of ordinary skill in the art in immediate possession of a catalytic process wherein the catalyst contains a binder. The present claims, by contrast, expressly require binder-free catalysts. Because Kushnerick et al. fails to disclose an alkylation process which utilizes the binder-free catalysts of the present claims, it is not a proper reference under 35 U.S.C. § 102(b). Accordingly, Appellant respectfully requests that the rejection of claims 1-6, 8, and 10 be reversed.

B. Kushnerick et al. Fails to Disclose the Transalkylation Process of Claims 7 and 9.

Claims 7 and 9 are deemed separately patentable over claims 1-6, 8, and 10, because claims 7 and 9 are directed to a transalkylation process. Kushnerick et al. does not disclose a transalkylation process, much less a transalkylation process, performed with the binder-free molecular sieves of the present claims. Accordingly, Kushnerick et al. cannot be said to anticipate claims 7 and 9, and the rejection should be reversed.

II. The rejection under 35 U.S.C. § 103(a) should be reversed.

Claims 1-10 are rejected under 35 U.S.C. § 103(a) as allegedly being unpatentable over Kushnerick et al. Appellants respectfully submit this rejection should be reversed.

A. Kushnerick et al. Fails to Render Obvious the Alkylation Process of Claims 1-6, 8, and 10.

Not only does the cited reference fail to teach or suggest the claimed invention, but one of ordinary skill in the art would have been led away from the alkylation process of the present claims. The Examiner states that Kushnerick et al. discloses inactive materials, functioning as diluents, which control the reaction rate. Final Office Action at page 3. The Examiner concludes that the artisan willing to make the trade-off between higher activity and better crush strength would omit a binder. Id. Appellants disagree with the Examiner's conclusion.

The relevant teachings of Kushnerick et al. to which the Examiner refers are as follows:

Inactive materials suitably serve as diluents to control the amount of conversion so that alkylation products can be obtained economically and orderly without employing other means for controlling the rate of reaction . . . [furthermore, it] is desirable to provide catalysts having good crush strength . . . [and] clay binders . . . are employed for the purpose of improving crush strength of the catalyst.

Col. 9, lines 29-42 (emphasis added).

It necessarily follows that one of ordinary skill in this art, upon viewing the passage relied upon by the Examiner, would be motivated to add a binder to the catalyst in order to improve the economic efficiency of the reaction and improve the crush strength of the catalyst. The claimed invention, however, results in a more efficient process in the

absence of such diluents or binders. This would not be expected from the teachings of the reference. The Examiner has pointed to no teaching or suggestion in this reference that would motivate one of ordinary skill in the art to conduct the alkylation process of the present claims in the presence of a binder-free crystalline molecular sieve. At the time of the invention, one of ordinary skill in the art would have to have been willing to sacrifice both an orderly reaction rate *and* crush strength in order to arrive at the claimed process. Based on the teachings of the reference, the ordinary practitioner would have been motivated to provide a catalyst comprising a clay binder. The fact that Kushnerick et al. only exemplifies bound catalysts solidifies this premise.

B. Kushnerick et al. Fails to Render Obvious the Transalkylation Process of Claims 7 and 9.

Claims 7 and 9 are deemed separately patentable over claims 1-6, 8, and 10, because claims 7 and 9 are directed to a transalkylation process. Kushnerick et al. fails to suggest *any* transalkylation process to one of ordinary skill in the art, much less the process of the present claims. Accordingly, Kushnerick et al. cannot be said to render obvious claims 7 and 9, and the rejection should be reversed.

C. The Evidence of Record is Sufficient to Rebut an Obviousness Rejection.

Assuming *arguendo* the Examiner has established a proper rejection under Section 103(a), and Appellants maintain no such rejection has been made, the showing of unexpectedly superior results in the specification is sufficient to rebut the rejection.

Appellants have found that alkylation and transalkylation products can be obtained most economically with catalysts that are self-bound, *e.g.*, catalysts without the binders of Kushnerick et al. It would not have been possible, except through the impermissible use of hindsight-based reasoning, to expect that the self-bound catalysts of the present claims would provide a reaction efficiency far-exceeding that of the prior art catalysts.

For the convenience of the Honorable Board, the evidence of unexpectedly superior results in the present specification is summarized here:

I. The catalysts

(a) Preparation of MCM-22/Al₂O₃ catalyst (Comparative catalyst)

The Al₂O₃-bound MCM-22 catalyst was prepared by mulling 65 parts of MCM-22 (the synthesis of which is described in U.S. Patent No. 4,954,325) and 35 parts of pseudoboehemite alumina in water. The mulled powder was formed into 1/16" extrudates using a screw-type extruder. After drying at 250°F, the extrudate was calcined in nitrogen at 900°F. After cooling to ambient air temperature, the extrudate was

exchanged with ammonium nitrate to remove residual sodium. After drying, the extrudate was calcined at 1000°F in air.

(b). Preparation of a binder-free MCM-22 catalyst (Inventive catalyst)

The binder-free MCM-22 catalyst according to the present invention was prepared by mulling 100 parts of MCM-22 crystals with water. The mulled powder was formed into 1/16" extrudates using a screw-type extruder. After drying at 250°F, the extrudate was exchanged with ammonium nitrate to remove residual sodium. After drying, the extrudate was calcined in nitrogen at 900°F followed by a calcination at 1000°F in air.

II. Comparison of catalyst performance for ethylbenzene synthesis

Figures 1 and 2 compare catalyst activity for liquid phase ethylbenzene synthesis at 500 psig, 5.5 benzene/ethylene molar ratio with temperature adjusted between 160 and 200°C and ethylene WHSV adjusted between 0.55 and 4.4 hr⁻¹.

Figure 1. Ethylbenzene Synthesis with Alumina-Bound MCM-22

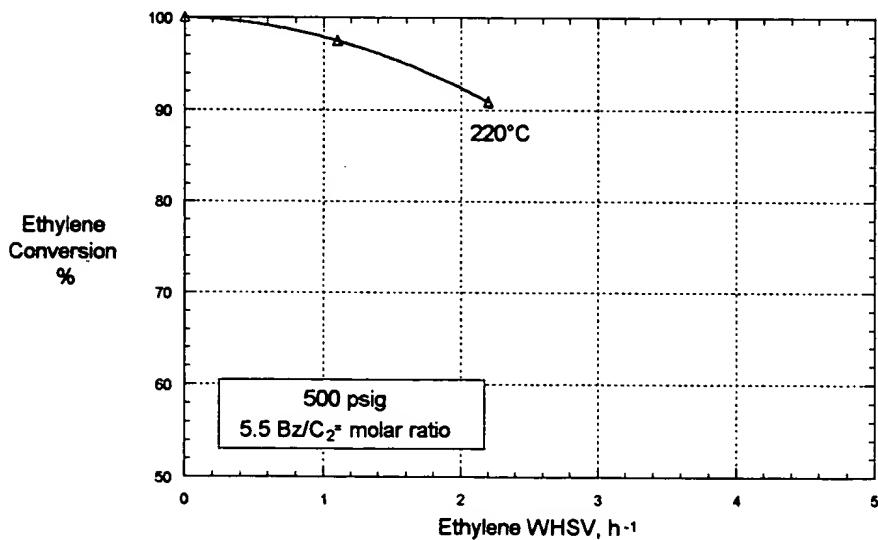
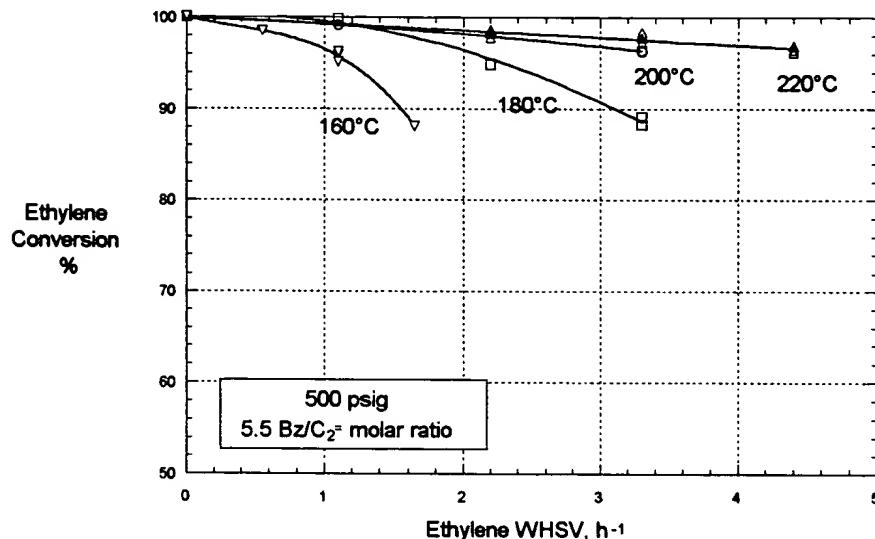


Figure 2. Ethylbenzene Synthesis with Self-Bound MCM-22

To achieve a constant ethylene conversion (e.g., 98%) at 220°C, the inventive binder-free MCM-22 could be operated at 3-4 times higher throughput than the Al₂O₃-bound MCM-22 catalyst of the reference. At constant throughput, *the binder-free MCM-22 provided ≥50°C temperature advantage over the Al₂O₃-bound MCM-22 to achieve comparable ethylene conversions.*

These results are significant. To be able to operate an ethylene conversion process with a ≥50°C temperature advantage results in a tremendous savings of energy. Such unexpectedly superior results could not have been expected from the teachings of Kushnerick et al.

The catalyst performances for liquid phase ethylbenzene synthesis are further compared in Table 2:

Table 2. Ethylbenzene Synthesis via Benzene/Ethylene Alkylation

Catalyst	Al ₂ O ₃ -bound MCM-22	Binder-free MCM-22	
Reaction Temperature, °C 220	220		161
Ethylene WHSV 4.4	1.1		1.1
Ethylene Conversion, % 96.9	97.4		96.7
Days on Stream 0.4	2.0		5.4
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Product Distribution, wt.%			
Ethylbenzene	93.66	93.36	90.82
Diethylbenzene	6.02	6.22	8.48
<u>Triethylbenzene</u>	<u>0.19</u>	<u>0.24</u>	<u>0.55</u>
Σ	99.87	99.82	99.85
Lights	0.03	0.01	0.00
Xylenes	0.00	0.00	0.00
n-C ₃ -Bz + Cumene	0.00	0.00	0.00
sec-C ₄ -Bz	0.06	0.10	0.08
<u>Heavies</u>	<u>0.04</u>	<u>0.07</u>	<u>0.08</u>
Σ (By Products)	0.13	0.18	0.16

To achieve comparable conversion at 1.1 ethylene WHSV, the binder-free catalyst could be operated at 161°C vs. 220°C with the Al₂O₃-bound catalyst. To achieve comparable conversion at 220°C, the binder-free catalyst could be operated at 4.4 ethylene WHSV vs. 1.1 ethylene WHSV with the Al₂O₃-bound catalyst.

Similarly results may been seen in Example 3 and Comparative Example 3, directed to liquid phase cumene synthesis *via* benzene-propylene alkylation over Al₂O₃-bound and binder-free MCM-22 catalysts, respectively. To achieve a constant propylene conversion (e.g., 96%) at 130°C average reactor temperature, the binder free MCM-22

catalyst could be operated at 100 propylene WHSV; by comparison, the Al₂O₃-bound catalyst operates at 40 propylene WHSV.

With regard to transalkylation processes, Comparative Example 4 and Example 4 on pages 19 and 20 of the specification show the unexpectedly superior properties of the claimed invention. The binder-free MCM-22 offered significantly higher conversions than the Al₂O₃-bound MCM-22. See Specification at Fig. 4 and Table 4.

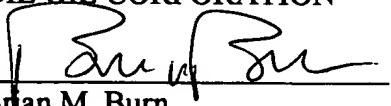
None of the unexpectedly superior results set forth in the specification could have been predicted by one of ordinary skill in the art. As a result, even if a proper Section 103(a) rejection had been established, the rejection would have been sufficiently rebutted by the evidence of record.

Conclusion

The Kushnerick et al. reference fails to anticipate or render obvious the claimed invention. Even if a *prima facie* case of obviousness were established, the data of record is sufficient to overcome the rejection. For the above reasons, it is submitted that the rejections should be reversed.

Respectfully submitted,

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